RAPID PUBLICATION

Microstructure of Zinc-Bonded $\text{Sm}_2\text{Fe}_{17}\text{N}_3$ Permanent Magnet Studied by Transmission and Analytical Electron Microscopy

Kenji Hiraga, Kensuke Okamoto[†] and Takahiko Iriyama^{††}

Institute for Materials Research, Tohoku University, Katahira, Aoba-ku, Sendai 980, Japan

Microstructure of the Zn-bonded $\text{Sm}_2\text{Fe}_{17}\text{N}_3$ permanent magnet having high magnetic coercivity of 11.8 kOe was studied by transmission and analytical electron microscopy. Three phases, α -Fe including 20 to 30 at%Zn, Fe_3Zn_7 , and $\text{Sm}_2(\text{Fe}, \text{Zn})_{17}\text{N}_x$, were found in addition to the matrix $\text{Sm}_2\text{Fe}_{17}\text{N}_3$ phase. The $\text{Sm}_2(\text{Fe}, \text{Zn})_{17}\text{N}_x$ phase, in which a fraction 0.2 of Fe in $\text{Sm}_2\text{Fe}_{17}\text{N}_3$ is replaced by Zn, surrounds the matrix grains and protects them from coming into contact with the α -Fe grains, which are considered to act as nucleation sites of reverse magnetic domains. It was concluded that the existence of the $\text{Sm}_2(\text{Fe}, \text{Zn})_{17}\text{N}_x$ phase causes the high coercivity.

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I. Introduction

Recently, the $\text{Sm}_2\text{Fe}_{17}\text{N}_3$ compound with excellent intrinsic magnetic properties was found by a group of Asahi Chemical Industry Co.⁽¹⁾, and independently by Coey and Sun⁽²⁾. Since the compound is prepared by nitrogenating powders of the $\text{Sm}_2\text{Fe}_{17}$ compound, it has been paid attention as a potential material for use as a bond-typed permanent magnet. Otani *et al.*⁽³⁾ found that zinc-bonding resulted in high coercivity for this magnet; the value of coercivity and maximum energy product of 0.66 T and 84 kJ/m³ was obtained in a Zn-bonded magnet with 15 vol%Zn. Also they found the formation of Fe_3Zn_7 in the $\text{Sm}_2\text{Fe}_{17}\text{N}_{2.3}$ matrix phase by X-ray diffraction. The purpose of the present work is to examine microstructure of the Zn-bonded permanent magnet and to clarify the reason why Zn-bonding establishes the high coercivity.

II. Experimental Procedures

The $\text{Sm}_2\text{Fe}_{17}\text{N}_3$ permanent magnet bonded with 30 wt%Zn metal was prepared by hot-pressing a mixture of $\text{Sm}_2\text{Fe}_{17}\text{N}_3$ and Zn powders at 450°C for 1 h. Magnetic coercivity of the magnet is 939.5 kA/m (11.8 kOe), which is higher than 3.4 kOe of row $\text{Sm}_2\text{Fe}_{17}\text{N}_3$ powder. Transmission electron microscopic observation and composition analysis were made with 200 kV electron microscopes (JEM-200CX, 2010-EX) equipped with an

energy dispersive X-ray spectroscopy (EDX). The microscope of 2010-EX can carry out EDX analysis with a minimum beam size of 2 nm. Analytical values by EDX are probably in error by as much as a few %, less than 5%.

III. Results and Discussion

Figure 1 is a transmission electron micrograph showing a typical microstructure near a boundary between two matrix $\text{Sm}_2\text{Fe}_{17}\text{N}_3$ grains. In Table 1, results of EDX analysis at various regions in the micrograph were listed up. From the data in the Table, one can see four phases with different compositions: the matrix $\text{Sm}_2\text{Fe}_{17}\text{N}_3$, $\text{Sm}_2(\text{Fe}, \text{Zn})_{17}\text{N}_x$, α -Fe primary solid solution, and Fe_3Zn_7 phases. The α -Fe primary solid solution includes 20 to 30 at%Zn, and the $\text{Sm}_2(\text{Fe}, \text{Zn})_{17}\text{N}_x$ is formed by replacing a fraction 0.2 of Fe in $\text{Sm}_2\text{Fe}_{17}\text{N}_3$ by Zn. The α -Fe grains have an averaged size of about 50 nm, and the Fe_3Zn_7 showing smooth contrast have a grain size of a few hundred nm. The $\text{Sm}_2(\text{Fe}, \text{Zn})_{17}\text{N}_x$ phase shows fine contrast and consists of micro-crystallines. Morphology of the four phases in Fig. 1 is schematically drawn in Fig. 2. The $\text{Sm}_2(\text{Fe}, \text{Zn})_{17}\text{N}_x$ phase is surrounding the matrix grains, and the α -Fe phase appears outside the $\text{Sm}_2(\text{Fe},$

Table 1 Compositions determined by EDX analysis at the A-J regions in Fig. 1.

	A	B	C	D	E	F	G	H	I	J
Sm (at%)	10	0	0	12	10	0	1	9	1	10
Fe	89	69	33	67	67	35	76	70	72	69
Zn	1	31	67	21	23	65	23	21	27	21

[†] Graduate Student, Tohoku University.

^{††} Central Laboratory, Asahi Chemical Industry Co., Ltd. Samejima, 2-1, Fuji 416, Japan.

Fig. 1 Transmission electron micrograph showing a typical microstructure of a grain boundary between two matrix grains. Letters, A-J, indicate the analytical regions with EDX.

Zn_{17}N_x , adjoining to the Fe_3Zn_7 grains. The Fe_3Zn_7 grains are present near the middle of the grain boundary. It should be noted that the $\text{Sm}_2(\text{Fe}, \text{Zn})_{17}\text{N}_x$ protects the matrix grains from coming into contact with the α -Fe grains, which have soft magnetic properties and are considered to act nucleation sites of reverse magnetic domains. Formation process of the morphology may be understood as follows. First, the reaction between Zn metal and $\text{Sm}_2\text{Fe}_{17}\text{N}_3$ grains occurs and produces the $\text{Sm}_2(\text{Fe}, \text{Zn})_{17}\text{N}_x$ phase that a part of Fe in the matrix are replaced by Zn. Consequently, an excess of Fe is ejected. The extra Fe and remained Zn make a binary equilibrium state of Fe-Zn; that is, the coexisting state of the α -Fe primary solid solution and the Fe_3Zn_7 phase. The α -Fe including 20 to 30 at% Zn shows the supersaturated solid solution of Zn, compared with a few % of the solid solution limit at 450°C. The last situation may be confirmed from the fact that any Zn-richer phases than Fe_3Zn_7 composition were not observed. In order to remove perfectly the α -Fe grains, a greater amount of Zn is required, as

the Zn-richer phases than Fe_7Zn_7 are formed.

It is worthwhile discussing the mechanism of high coercivity from the microstructure obtained by the present work. It was considered so far that the high coercivity of this Zn-bonded permanent magnet was established by removing an impurity Fe phase, remained in the preparation process of $\text{Sm}_2\text{Fe}_{17}$, by the formation of Fe_3Zn_7 . However, the present observation shows that such simple reaction does not occur. The reaction between the matrix phase and Zn metal produces the $\text{Sm}_2(\text{Fe}, \text{Zn})_{17}\text{N}_x$ phase and the excess Fe. A main part of the excess Fe are spent by the formation of the Sm_2Zn_7 phase, but part of Fe always remain as the α -Fe-Zn primary solid solution of a coexisting phase with the Sm_2Zn_7 phase. However, the α -Fe phase, which is considered to deteriorate magnetic coercivity of the permanent magnet, is intercepted to touch to the matrix phase by the $\text{Sm}_2(\text{Fe}, \text{Zn})_{17}\text{N}_x$ phase surrounding the matrix grains. That is, the $\text{Sm}_2(\text{Fe}, \text{Zn})_{17}\text{N}_x$ phase blocks the invasion of reverse domains, born at the α -Fe grains, upon the matrix grains. Also, the

Fig. 2 Schematic illustration of the microstructure in Fig. 1, showing regions of $\text{Sm}_2\text{Fe}_{17}\text{N}_3$ (O), $\text{Sm}_2(\text{Fe}, \text{Zn})_{17}\text{N}_x$ (P), $\alpha\text{-Fe}$ (Q) and Fe_3Zn_7 (R) phases.

$\text{Sm}_2(\text{Fe}, \text{Zn})_{17}\text{N}_x$ phase produces smooth surfaces of the matrix grains, which remove weak nucleation sites of reverse domains at surfaces. The above idea for the high coercivity mechanism of this magnet is based on the assumption that the $\text{Sm}_2(\text{Fe}, \text{Zn})_{17}\text{N}_x$ phase is a non-magnetic material. The measurement of magnetic properties of the $\text{Sm}_2(\text{Fe}, \text{Zn})_{17}\text{N}_x$ is an urgent necessity.

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